

IX. OTHER IMPACTS

Multiuser Spectroscopic Facility Development

Development in the multiuser spectroscopic facility has focused on two areas: hyper-Rayleigh scattering and optical microscopy. Progress in each area is discussed below:

Hyper-Rayleigh Scattering. Hyper-Rayleigh Scattering (HRS) provides a direct measure of molecular first hyperpolarizability (β). This technique is complimentary to the more common technique of Electric Field Induced Second Harmonic Generation (EFISH).³³ In EFISH, an applied electric field induces asymmetry in the system such that second-harmonic generation of an incident laser field occurs. The magnitude of the second-harmonic field is dependent on both molecular dipole moment (μ) and β . To extract β from an EFISH measurement, both μ and β must be known. In a HRS experiment, monochromatic light with frequency ω is incident on a sample and the scattering at 2ω is observed, referred to as the hyper-Rayleigh. The intensity of scattering at 2ω is diagnostic of the magnitude of β . As such, HRS provides a more direct methodology for measuring molecular hyperpolarizabilities.

Although, HRS is a promising method for quantifying molecular non-linear properties, the primary issue limiting the applicability of this technique was that of wavelength tunability. Much of the previous HRS work was performed using 1.9 μm light from a high-powered hydrogen-shifted Nd:YAG laser. Although these experiments were groundbreaking, they were limited to 1064 nm and 1906 nm. The limitations of Nd-YAG excitation sources can in theory be avoided by the use of higher-repetition rate sources, in particular Ti:Sapphire oscillators. However, these systems are tunable over a limited wavelength range (700 nm to \sim 1000 nm) such that the measurement at wavelengths relevant to telecommunications applications (1.3 μm and 1.5 μm) can not be performed. Only recently have such wavelengths been available through the use of tunable IR optical parametric oscillators.

To address the limitations of excitation tunability outlined above, we have constructed a HRS apparatus based on a Ti:sapphire pumped optical parametric oscillator (Figure 1). First, the output of a Ti:Sapphire oscillator (Spectra Physics Tsunami) can be used for excitation from 740 to 1000 nm. Further tunability is achieved by using the oscillator to pump an optical parametric oscillator (Spectra Physics Opal) which provides output ranging from 1.1 μm to 2.5 μm . This system is now operative, and has been used to provide estimates of β for approximately 16 chromophores recently developed by researchers involved in the STC.

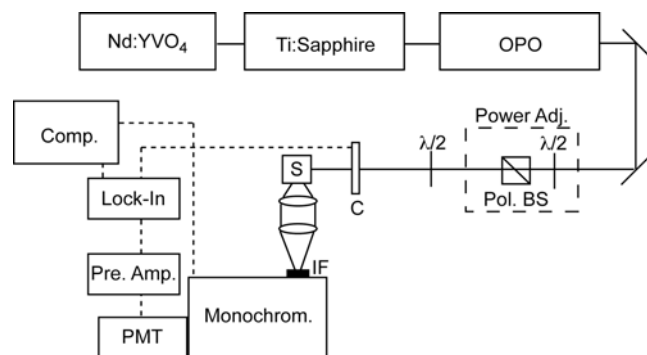


Figure 1. Schematic representation of the HRS apparatus. Excitation is performed using either the Ti:Sapphire oscillator or optical-parametric oscillator (OPO) output. Power is adjusted using a half-wave plate ($\lambda/2$) and polarizing beamsplitter (Pol. BS.). The excitation beam is modulated using a mechanical chopper (C) synchronized to a lock-in for phase-sensitive detection. The HRS scattering is collected from the sample (S) and delivered to a monochromator. Signal from the PMT is delivered to a pre-amplifier before being input to the lock-in.

Optical Microscopy. To characterize large assemblies of non-linear optical chromophores doped into polymeric systems, we are developing both linear and non-linear optical microscopy facilities. This facility is used by researchers in the STC to characterize materials, and by students participating in the educational and training mission of the STC. A schematic of the two-photon optical microscope we have constructed and that is currently operational is presented in Figure 2. Excitation is accomplished using a tunable femtosecond Ti-sapphire oscillator (home built) pumped by a Nd:YVO₄ laser (Millennia, Spectra Physics). The oscillator is tunable from 750 nm to 850 nm with a temporal pulse width of 30 fs (full width at half maximum). Before entering the microscope, the excitation beam diameter is increased to fill the microscope objective aperture, with adjustment of the incident power performed using a zero-order half-waveplate in combination with a thin-film polarizer. A second waveplate is used to adjust the polarization of the incident light. An 800-nm dichroic beamsplitter directs the excitation beam through a 1.3-NA oil-immersion objective (Nikon) to a diffraction-limited spot of ~400-nm in diameter. Positioning of the objective relative to the sample is controlled using a piezo-electric actuator (ThorLabs). The sample is scanned via an XY piezo-electric stage (Queensgate) having a spatial range of 100 μm and resolution of 10 nm. Emission is collected by the objective, returns through the dichroic beamsplitter, and is focused using a 200-mm achromatic lens. Rejection of residual fundamental is accomplished using a 720-nm short-pass filter. The emission can be directed to a CCD camera for alignment, or imaged onto a pair of single-photon counting photomultiplier tubes (Hamamatsu HC135-01) that are not responsive at the excitation wavelengths employed. An avalanche photodiode/photon counting module (Perkin Elmer) is also available for use. As the figure illustrates, polarization optics are available to measure the linear dichroism of the fluorescence in order to determine molecular alignment (see below). We will be taking delivery of a confocal microscope in the upcoming month, and will be installing this system in early summer.

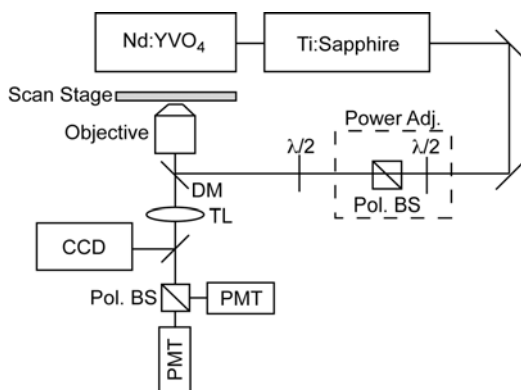


Figure 2. Schematic of the two-photon optical microscope. $\lambda/2$ = half-wave plate, DM = dichroic mirror, TL = tube lens, Pol. BS = polarizing beamsplitter, PMT. Other details are provided in the text.